



Full Length Article

Measurements of sooting limits in laminar premixed burner-stabilized stagnation ethylene, propane, and ethylene/toluene flames

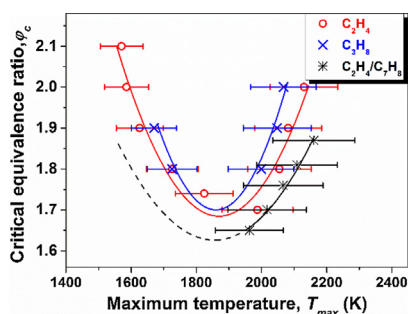
Quanxi Tang^{a,b}, Mengda Wang^{a,b}, Xiaoqing You^{a,b,*}

^a Center for Combustion Energy, Tsinghua University, Beijing 100084, China

^b Key Laboratory for Thermal Science and Power Engineering of Ministry of Education, Tsinghua University, Beijing 100084, China



GRAPHICAL ABSTRACT



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ABSTRACT

The systematic investigation of the effect of two major experimental parameters (equivalence ratio and temperature) as well as fuel structure on the sooting limits of ethylene, propane, and ethylene/toluene flames was carried out in premixed burner-stabilized stagnation flames through qualitatively visual observations of flame yellow luminosity and quantitatively direct measurements of incipient soot particle size distributions by a scan mobility particle sizer. A strong temperature dependence of incipient soot formation was observed in the three flames. Detailed chemical kinetic modeling of the flame structure and the soot precursor chemistry was performed to explain the experimental observations. The results show that the incipient soot formation is prohibited in either low or high temperature flames. While the former is due to slow reactions induced by both low temperatures and small concentrations of soot precursors, the latter is attributed to the thermodynamic reversibility of polycyclic aromatic hydrocarbons (PAHs) at high temperatures. Sooting limits have been found to be not a function of concentrations of PAHs alone at a fixed flame temperature; sooting propensity generally increases with increasing fuel carbon/hydrogen atom ratio, presenting the following order: propane < ethylene < ethylene/toluene.

1. Introduction

Sooting tendencies of individual hydrocarbons can be used to estimate the effects of fuel composition on soot inception [1], to introduce the effects of fuel structure into soot models [2], to assess the effect of

C/H and fuel structure on soot formation [3,4], as well as for many other purposes [5]. The means to assess sooting tendencies include: (1) the equivalence ratio at sooting limits (i.e. the critical equivalence ratio where yellow flame luminosity just begins to appear) [3]; (2) the threshold sooting index (TSI) that depends on the heights of jet flames

* Corresponding author at: Center for Combustion Energy, and Department of Energy and Power Engineering, Tsinghua University, Beijing 100084, China.

E-mail address: xiaoqing.you@tsinghua.edu.cn (X. You).

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at the smoke point [4,6–9]; (3) the yield sooting index (YSI) that relates to the maximum soot volume fraction on the centerline of a coflow flame [5,10]. The above-mentioned first method is traditionally used to determine the critical equivalence ratio φ_c -maximum temperature T_{\max} map for premixed systems, which can be utilized to optimize internal engine design [11,12], especially in the low temperature range achieved by adding high levels of exhaust gas recirculation [13]. However, regions of soot inception are uncertain for the soot- φ_c - T_{\max} maps [11]. As such, more quantitatively experimental measurements in well-controlled configurations are essential for establishing more reliable soot- φ_c - T_{\max} models.

Based on the sooting limit measurements from Bunsen flames, in a pioneering investigation by Street and Thomas [14], the sooting tendencies of different types of fuel were ranked as aromatics > alcohols > alkenes > acetylene. However, the influence from flame temperature was neglected in the experimental results. In order to determine the fuel structure effect at a fixed flame temperature, Takahashi and Glassman [3] conducted a systematic series of experiments on sooting limits in a Bunsen-type burner with special attention to controlling the flame temperature by varying the concentration of nitrogen. As expected, the ranking of φ_c at a fixed temperature was different from that in [14], as φ_c (acetylene) < φ_c (ethene) < φ_c (ethane). Takahashi and Glassman [3] concluded that sooting limits were mainly related to the competing reaction rates of fuel pyrolysis and oxidative attack, which were strongly dependent on the flame temperature.

It has been widely accepted that the flame temperature has a strong effect on sooting limits but in a more complicated way [15–17]. A consensus has been reached that sooting limits follow a fall-then-rise or U-shaped behavior with the increase of flame temperature [17–19]. The nature behind the experimental observations, however, is still in debate. In the early studies, researchers suggested the concentration of OH was closely correlated with flame temperature, thus determining the concentration of soot precursors via the oxidation by OH [20]. Markatou et al. [21] suggested that the decreasing concentration of PAH is mainly attributed to thermodynamic reversibility towards high temperatures, while oxidation behavior by OH due to a high flame temperature played a more important role on light hydrocarbons (such as C_2H_3), rather than PAH.

For reasons discussed above, it would be beneficial to provide more quantitative experimental data of sooting limits in a well-defined configuration for evaluating the accuracy of soot models, especially in the low temperature range, where the experimental data, to the best of our knowledge, are quite scarce. The traditional method for determining sooting tendencies is based on TSI or YSI with optical diagnostic techniques, which can assess sooting tendencies of hundreds of individual hydrocarbons in high-temperature regions [4,5,10,22,23]. However, they are not the best fit for the studies on sooting limits in low-temperature regions. In this work, the sooting limit is determined by both the qualitatively visual observation and the quantitative measurement of particle size distribution functions (PSDFs) with a scan mobility particle sizer (SMPS) in a series of burner-stabilized stagnation (BSS) flames. Having been proved to be as sensitive as laser induced incandescence to detect the onset of soot formation [24], the chosen SMPS was used to investigate sooting limits due to its capability to provide spatial distributions of nascent soot which could be used to validate model assumptions [25–28]. Based on this method, we carried out a systematic experimental study on sooting limits of different fuel types (alkane, olefin, aromatics) over a wide range of flame temperatures (1500–2200 K) and equivalence ratios (1.6–2.1). In our study, BSS flames are chosen for the simple flame structure (pseudo one-dimensional distribution) that helps us quantify flame temperature profiles more directly. In the meanwhile, results on the operating parameter effects of this experimental setup are also made known [29]. Compared with those flames in jet and Bunsen-type burners [3,5], sooting limits at lower temperatures can be achieved in the BSS flames due to the water-cooled porous plug in the McKenna burner and the water-cooled

stagnation plate. In addition, we also used detailed chemical kinetic modeling to gain insight of the gas-phase reactions.

2. Experimental and computational methods

Laminar premixed flames were generated by a commercial McKenna burner. Liquid fuels were fully vaporized using a fuel vaporization system [30]. Soot samples were then drawn into a micro-orifice embedded within and flush to the stagnation surface on a water-cooled aluminum disc to be analyzed by a scanning mobility particle sizer (TSI 3936). The detailed description of the experimental setup and procedure can be found in our previous studies [29–31].

Particle-particle coagulation is one of the major causes of particle losses in the sampling line, which can be minimized by optimal dilution using nitrogen. The adopted procedure for determining optimal dilution ratio was from previous studies [29,32]. For the current setup, the optimal dilution ratio is between 1500 and 5000, a range to within which PSDFs are insensitive to any change in dilution ratios. Parameterized correction of mobility particle size was adopted according to theoretical formula introduced in Ref. [33]. All soot particle sizes reported hereafter are corrected mobility diameters.

To measure the axial flame temperature profiles, an S-type thermocouple coated with Y/Be/O mixture was used to prevent catalytic reactions on the surface of thermocouple. The diameter of a thermocouple before and after coating is 125 and 142 μm , respectively. The detailed description of radiation corrections to the measured temperature and the analysis of uncertainties in the temperature measurements can be found in our previous work [30].

Table 1 summarizes the flame conditions for determining sooting limits. The sooting limits of three series of flames, C_2H_4 (E series), C_3H_8 (P series), and $C_6H_5CH_3/C_2H_4$ (T series) are assessed over a wide range of temperatures and equivalence ratios. For the measurements of sooting limits of all fuels, the burner-to-stagnation surface separation distances are 1.0 cm at the low temperature branch and 2.0 cm at the

Table 1
Summary of the flame conditions^a for determining sooting limits.

| Flame | Equivalence ratio, φ | Compositions | Cold gas velocity, $v_0(\text{cm/s})^c$ | $T_{\max}(\text{K})^b$ |
|-------|------------------------------|--------------|---|---|
| — | — | C_2H_4 | O_2 | — |
| E1 | 2.1 | 0.148 | 0.212 3.4 | $H_p = 1.0 \text{ cm}^d$ 1571 ± 66 |
| E2 | 2.0 | 0.144 | 0.216 3.6 | 1586 ± 68 |
| E3 | 1.9 | 0.140 | 0.220 3.7 | 1626 ± 72 |
| E4 | 1.8 | 0.135 | 0.225 4.5 | 1727 ± 80 |
| E5 | 1.74 | 0.132 | 0.228 6 | 1825 ± 89 |
| — | — | C_3H_8 | O_2 | — |
| E6 | 1.7 | 0.135 | 0.225 10 | $H_p = 2.0 \text{ cm}$ 1988 ± 93 |
| E7 | 1.8 | 0.140 | 0.220 13 | 2055 ± 97 |
| E8 | 1.9 | 0.144 | 0.216 15 | 2082 ± 102 |
| E9 | 2 | 0.148 | 0.212 17 | 2131 ± 104 |
| — | — | $C_6H_5CH_3$ | O_2 | — |
| P1 | 1.9 | 0.099 | 0.261 2.8 | $H_p = 1.0 \text{ cm}$ 1670 ± 69 |
| P2 | 1.8 | 0.095 | 0.265 3.2 | 1726 ± 76 |
| — | — | C_2H_4 | O_2 | — |
| P3 | 1.8 | 0.095 | 0.265 8.5 | $H_p = 2.0 \text{ cm}$ 1999 ± 101 |
| P4 | 1.9 | 0.099 | 0.261 10.5 | 2048 ± 104 |
| P5 | 2.0 | 0.103 | 0.257 13.5 | 2068 ± 101 |
| — | — | C_3H_8 | O_2 | — |
| T1 | 1.65 | 0.0643 | 0.0276 0.268 4.5 | $H_p = 1.0 \text{ cm}$ 1963 ± 104 |
| T2 | 1.70 | 0.0659 | 0.0283 0.266 5.3 | 2018 ± 122 |
| T3 | 1.76 | 0.0675 | 0.0289 0.264 6.5 | 2067 ± 122 |
| T4 | 1.81 | 0.0690 | 0.0296 0.261 8.7 | 2109 ± 124 |
| T5 | 1.87 | 0.0705 | 0.0302 0.259 11.9 | 2161 ± 126 |

^a Unburned gas composition: Fuel- O_2 -0.64 Ar (by mole).

^b T_{\max} is the measured maximum flame temperature with radiation correction.

^c STP condition (298 K and 1 atm).

^d H_p is the burner-to-stagnation surface separation distance.

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